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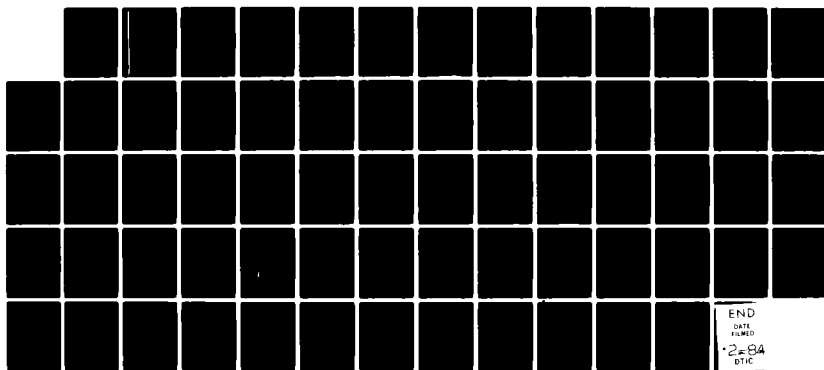
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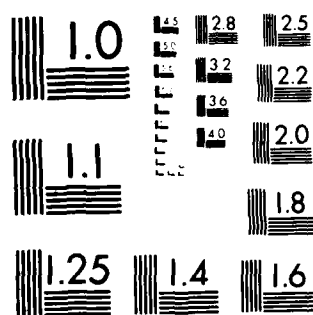
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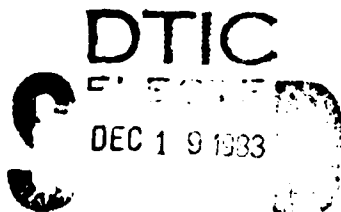


THE UNIVERSITY OF NEW MEXICO  
COLLEGE OF ENGINEERING

# BUREAU OF ENGINEERING RESEARCH

Final Report

Optical Thin Film Workshop



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by

Kenneth Jungling

April 21-23, 1982

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Final Report

Optical Thin Film Workshop

Kenneth Jungling  
Electrical and Computer Science Department

and

Institute for Modern Optics

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Workshop Advisory Committee

A. Guenther	Air Force Weapons Laboratory
K. Jungling	University of New Mexico (Chairman)
J. McNeil	University of New Mexico (Co-chairman)
L. Myers	Air Force Office of Scientific Research
B. Pierce	Air Force Weapons Laboratory
H. Winsor	Air Force Office of Scientific Research

## ABSTRACT

A three-day workshop on optical thin films was conducted on 21-23 April 1982 by the University of New Mexico for the Air Force Office of Scientific Research. The objective of this workshop was to generate a list of basic research efforts which should be pursued in thin film processing techniques and thin film diagnostics which would lead to the development of more damage resistant, more predictable and repeatable thin film coatings which would exhibit long life and stable performance. A group of invited papers covering laser damage, surface preparation, film microstructure, deposition techniques and diagnostics were presented. Discussion of the research needs to advance basic understanding and improvement of optical thin films is summarized, and a prioritized research program which has been generated by the Workshop Advisory Committee is given.



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## INTRODUCTION

Optical performance and laser damage are the principal design constraints in most high power/high energy systems. In order to satisfy system performance requirements, thin film coatings are applied to high energy laser (HEL) components such as mirrors, windows, or beam sampling devices. The principal functions being performed by coatings are (1) protective overcoats, (2) enhance reflections or (3) antireflection coatings, (4) wavelength filtering, (5) polarization selection, and (6) phase control.

Current deposition techniques do not produce predictable, repeatable or adequately durable, thin films. Although there has been much written concerning thin film technology, deposited state-of-the-art films do not exhibit parent (bulk) material properties, and laser damage thresholds are generally orders of magnitude lower than the intrinsic bulk material damage thresholds. The Institute for Modern Optics, University of New Mexico conducted a three-day thin film workshop for the Air Force Office of Scientific Research on 21-23 April 1982. Sixty-one people from universities, industry, and government participated. The disciplines represented were surface chemist and analysts, thin film deposition experts, material physicist, thin film characterization experts, laser damage specialists, and thin film microstructure analysts. The list of participants appears in Appendix A, while the agenda of the workshop appears in Appendix B. The overview paper "Laser Induced Damage in Optical Thin Films" presented by Dr. Guenther and the abstracts of the other presentations are in Appendix C. The objective of the workshop was to generate a prioritized list of basic research efforts which should be pursued in thin film processing techniques and thin film diagnostics leading to the development of more damage resistant, more predictable and repeatable thin film coatings. During the workshop the following activities were accomplished:

- (1) Pertinent thin film data were reviewed.
- (2) Thin film processing techniques were reviewed that included

- a. Substrate material and polishing procedures
- b. Substrate cleaning
- c. Substrate temperature and heating/cooling rates
- d. Evaporant materials and purity
- e. Deposition methods and rates
- f. System gas pressure and composition were reviewed.

(3) The requirements for instrumentation to provide better control of process parameters and more thorough thin film characterization were discussed.

(4) A review of laser induced damage processes was presented.

Presentations were made during the first 1 1/2 days. The next half day the group was divided into five working subgroups to brainstorm what basic research should be performed in the following areas:

- (1) Deposition methods for control of microstructure
- (2) Structure of thin film (optical/mechanical properties, homogeneity, etc.)
- (3) Microanalytical diagnostics
- (4) Substrate surface preparation and cleaning
- (5) Structure/optical properties that relate to damage
- (6) Optimum laser damage testing

A plenary session was conducted Friday morning so that each group's recommendations could be presented and discussed by all the attendees.

## PRIORITIZATION OF OPTICAL THIN FILM RESEARCH

Obtaining an understanding of optical thin films requires a multi-faceted, multi-discipline approach which is thoroughly planned and coordinated on a long term basis. Additionally, there is considerable research in semiconductor thin films and thin film characterization which may be directly applicable in advancing the understanding of optical thin films and laser induced damage mechanisms. To obtain, most efficiently, an understanding of optical thin films such that new processing techniques can be developed which will provide more damage resistant, predictable, durable and reproducible thin film coatings which would exhibit long life and stable performance, the establishment of a technical coordination/advisory council was given the highest priority for an optical thin film research program. The council would:

- 1) facilitate communication and transfer of existing technology between the many disciplines through sponsorship of conferences, workshops, review articles in appropriate journals, etc.
- 2) evaluate all existing macroanalytical and microanalytical diagnostic techniques and organize a program which may lead to the detection of subtle differences in coatings. Also it would encourage the development of new characterization techniques which could lead to a more comprehensive understanding of the total thin film arena from design to ultimate failure. This effort would evolve into the standardization of optical thin film evaluation techniques thus enabling expanded standards for high power/high energy optical thin film.
- 3) propose research in new areas and in new directions which will build the understanding of the physics, chemistry, structure, and their interrelation to optical thin films.

The second highest priority was to establish a framework for fundamental understanding of nucleation and growth under non-equilibrium

conditions. In many films there appears to be a 20-30Å microstructural unit. The complex relationship between the detailed physical structure of the film and the chemical, mechanical, optical and electronic properties is essentially unknown or very limited. Studies of nucleation and growth of thin films essentially were performed when diagnostic techniques were very limited and levels of material purity and surface cleanliness were not attainable as compared to the levels obtainable today. Additionally, film structure classification which includes the structural zone model and deposition phase diagrams should be actively pursued.

The next priority was to develop an understanding of structure and composition of the thin film at the substrate-coating interface or at film-film interfaces. Some interface states that should be characterized are the metal-film, film-film, film-water, and film-air. An understanding of diffusion in and across films and interfaces as well as diffusion of water and other atmospheric gases through the films needs to be developed. The role stress corrosion plays in laser induced damage and long-term stability needs to be established because many materials are under stress when in thin film form. Also, the surface chemistry that occurs when a material nucleates on another substance needs to be thoroughly examined; for example, when Si is deposited on  $\text{Al}_2\text{O}_3$ , a thin layer of  $\text{SiAl}_2\text{O}_3$  is formed.

The fourth priority stresses the characterization and distribution of defects and impurities in thin films (micro-characterization). When optical thin films are laser damage tested to determine their threshold, damage generally occurs at isolated,  $<10\text{ }\mu\text{m}$  size spots and subsequently grows. Presently no non-destructive characterization technique can predict at what level or location that laser induced damage will occur when the optical thin film is tested. Characterization after destructive laser damage testing has very limited usefulness because the thin film defects (inclusions, impurities, structural imperfections, etc.) have been highly distorted or removed.

It is imperative that the new diagnostic techniques such as sub-micron ellispometry, submicron SIMS using a laser beam evaporation, submicron fluorescence, etc., be applied as a systematic, logical approach to obtain a correlation between actual laser damage levels and characterization techniques. Special diagnostic techniques which can detect low levels of impurity, their distribution and chemical composition should be actively pursued.

Novel coating processes which include coating depositions in space have been placed in the fifth priority. Any new or evolving processes must be evaluated for the chance of producing a quantum jump (if there are any) in film quality. There was no question that some resources need to be placed in this category in any comprehensive program. However, the consensus was that a research program to obtain basic understanding of the thin films was the best way to obtain more damage resistant, predictable, durable, and reproducible thin film coatings. The many variables need to be parametrically varied in a controlled systematic fashion to determine the effect each variable has on damage threshold, stable performance, etc.

The sixth priority is to investigate the correlation between structural, optical, mechanical, absorption, and scattering properties and laser damage thresholds (macro-characterization). If a correlation exists between a macro-characterization technique and laser damage, it would be much simpler, faster and cost effective to implement. Large energy/high power, large spot, laser damage studies are very expensive and the data is not timely enough to have the greatest impact on coating development. For the test data to have the greatest impact, it should be available one to two weeks after the deposition has occurred and the vacuum chamber has not been utilized for other coating material depositions. It is imperative that a valid characterization be established which can provide the timely data required for thin film coating

development. The key to determining a relevant characterization technique is that a very systematic study between characterization techniques and laser damage studies needs to be performed. Present programs in optical thin film development do not provide for this because of time constraints and resource limitations.

The final priority is to determine the influence of the substrate and substrate surface characteristics on the deposited thin film. How do surface states affect nucleation? Does sub-surface damage affect nucleation, diffusion, etc.? Can surface modification through polishing or other treatments enhance adhesion and modify the thin film structure? What other thermomechanical considerations are important in any temperature cycling. These are some of the questions that should be addressed in this category.

The various priorities for optical thin research are now compiled in Table I.

Priority	Research
1.	Establish a technical/advisory council.
2.	Establish a framework for fundamental understanding of nucleation and growth under non-equilibrium conditions.
3.	Develop understanding of structure and composition thin films. The various interfaces, and the role diffusion plays.
4.	Micro-characterization and distribution of defects and impurities in thin films.
5.	Pursue novel coating techniques.
6.	Macro-characterization of thin films and how it correlates with laser damage.
7.	Evaluate the influence of the substrate characteristic on optical thin films.

It cannot be overemphasized that optical thin film research is a multi-faceted problem requiring close cooperation and open communication between the people performing the depositions, characterization, laser damage testing, and the theory. Such coordination and cooperation will insure progress and greatest return on the investment.



Appendix A

Attendance List  
Optical Thin Film Workshop

April 21-23, 1982

<u>NAME</u>	<u>AREA OF INTEREST</u>	<u>TELEPHONE NO.</u>
Joseph H. Apfel OCLI 2789 Northpoint Pkwy. Santa Rosa, Ca 95402		707-545-6440 OCLI 707-525-7184 APFEL
Milton Birnbaum Aerospace Corp. P.O. Box 92957 Los Angeles, CA 90009		213-648-6839
Troy W. Barbee, Jr. Dept. of Mat'ls Sci. & Eng. Stanford University Stanford, CA 94305	Material Synthesis X-Ray Optics Fracture Structural Characterization	415-497-1919
H. E. Bennett Michelson Lab, Code 38101 Naval Weapons Center China Lake, CA 93555		714-939-2869
Jeff Brinker Sandia National Laboratory Division 5845 Albuquerque, NM 87115	Sol-gel coatings	505-846-3552
J. D. Doll Los Alamos Nat. Lab CNC-2/MS-6738 Los Alamos, NM	Surface Dynamics/ surface self-diffusion	505-667-4686
Terry Donovan Naval Weapons Center China Lake, CA 93555	Thin Film Deposition, Diagnosis, Laser-Radiation Damage	714-939-3852
Richard Esposito Developmental Optics Fac. KAFB, Albuquerque, NM	Optical Coatings	505-844-1064
Graham Flint Developmental Optics Fac. Bldg. 400 KAFB West Albuquerque, NM 87119		505-844-1064

<u>NAME</u>	<u>AREA OF INTEREST</u>	<u>TELEPHONE NO.</u>
A. H. Guenther AFWL/CA Kirtland AFB Albuquerque, NM 87117		505-844-9856
T. W. Haas AFWL/MLBM Wright-Patterson AFB Ohio 45433	Research group leader for Surface and Interface Phenomena group	513-255-5892
Richard W. Hoffman Dept. of Physics Case Western Reserve Univ. Cleveland, OH 44106	Structure-property relations in thin films, especially mechanical & magnetic prop. stress, adhesion	216-368-4012
Samuel Holmes Northrop Res. & Tech. Palos Verdes, CA 90274	Optical thin films and deposition techniques	213-377-4811
Michael R. Jacobson Optical Sciences Center University of Arizona Lab. Tucson, AZ 85721		602-626-2382 Off. 602-626-2864 Sec. 602-626-3530
Kenneth Jungling University of NM/EECE Albuquerque, NM 87131	Optical Coatings Diagnostics	505-277-3317 Off. 505-277-2436 Secy.
Michael L. Knotek Division 5114 Sandia National Laboratories Albuquerque, NM 87185	Surface analysis (H & OH)	505-844-2272
W. J. Lackey Oak Ridge National Laboratory Oak Ridge, TN 38930 Stanford University Stanford, CA 94305		615-574-4551 FTS 624-4551
Ken Lakin 215 Applied Sciences Center Ames Laboratory Ames, IA 50011	Thin film growth & charac- terization AlN, SiO <sub>2</sub> , Al <sub>2</sub> O <sub>3</sub> , ZnO - Nondestructive evalua- tion - laser induced sound generation	515-294-7732

<u>NAME</u>	<u>AREA OF INTEREST</u>	<u>TELEPHONE NO.</u>
Ingolf Lindau Stanford Electronics Lab. Stanford University Stanford, CA 94305	Surface/Interface analysis & characterization	415-497-1052
Ronald L. Lusk AFWL/ARAO Kirtland AFB Albuquerque, NM 87117	User - all aspects - especially laser damage testing	505-844-1776
Myron T. Maclin AF Weapons Lab (AFWL/ARAO) Kirtland AFB Albuquerque, NM 87117	User-interested in all aspects of improved optical coatings	505-844-1776
Donald M. Mattox Sandia National Laboratory Division 5834 Albuquerque, NM 87115	Surface cleaning, thin films	505-844-7777
Keith McDowell LANL (USM) & Clemson Univ. (Chem.) Los Alamos, NM 87545 CNG2 G738		505-667-4686
John McIver Dept. Physics & Astronomy University of New Mexico Albuquerque, NM 87131	Laser damage	505-277-5909
J. R. McNeil Department of Elec. Eng. Institute for Modern Optics University of New Mexico Albuquerque, NM 87131	Optical thin film deposition & structure	505-277-5626
R. P. Merrill 201 Olin Hall Ithaca, NY 14650	Surface chemistry & physics	607-256-3854
Russell Messier Penn State University 265 Materials Research Laboratory University Park, PA 16802	Microstructure of thin films	814-865-3704

<u>NAME</u>	<u>AREA OF INTEREST</u>	<u>TELEPHONE NO.</u>
David Milam Lawrence Livermore Labs P.O. Box 5508 L-470 Livermore, CA 94550	Laser damage Laser diagnostics	415-422-5498
Lee E. Myers AFOSR/NC Bldg. 410 Bolling AFB, DC 20332		202-767-4963 297-4963 - Autovon
David J. Nagel Naval Research Lab - 6680 Washington, DC 20375	Surface diagnostics	202-767-2549
Yoshiharu Namba Dept. of Physics & Astronomy University of New Mexico Albuquerque, NM 87131	Polishing and surface analysis	505-277-5625
Vaidya Nathan AFWL/ARAO Kirtland AFB Albuquerque, NM 87117	Laser damage theory	505-844-3982
R. C. Pastor Hughes Research Laboratories 3011 Malibu Canyon Road Malibu, CA 90265	Material processing	213-456-6411
Walter T. Pawlewicz Battelle-Pacific Northwest Lab Richland, WA 99352	Sputtering, microstructure properties	509-373-2537 FTS 440-2537
Bruce J. Pierce AFWL/ARAO Kirtland AFB Albuquerque, NM 87117	User-interested in all aspects	505-844-1776
Tilak Raj International Laser Systems P.O.Box 9316 Albuquerque, NM 87119	Surface & thin film analyst	505-844-1064
Mehmet Rona Arthur D. Little, Inc. Acorn Park Cambridge, MA 02140	MBE	617-864-5770-x2534

<u>NAME</u>	<u>AREA OF INTEREST</u>	<u>TELEPHONE NO.</u>
Earl Rudisill Laser Power Optics San Diego, CA 92121		714-455-0751
David Sanders 8328 Bld. 223 National Bureau of Standards Washington, DC 20234	Glass thin films	301-921-2817
Howard R. Shanks A207 Phys. Bldg. Ames Laboratory Ames, IA 50011	Surface physics amorphous Si, SiC RF sputtering	515-294-6816
Robert W. Springer CMB-6, MS770 Los Alamos National Lab Los Alamos, NM 87545	Surface analysis, coatings	505-667-4258
Alan F. Stewart AFWL/ARAO Kirtland AFB Albuquerque, NM 87117	Laser damage, nondestructive	
Chater Stinespring Aerodyne Research, Inc. 45 Manning Park The Research Center at Manning Park Billerica, MA 01821	XPS/AES Characterization of Adsorbed species- bonding-sticking probabilities & surface segregation	
Paul Temple Code 3816 Naval Weapons Center China Lake, CA 93555	Laser Calorimetry-(surface vs. bulk absorption in films), laser damage, CO <sub>2</sub> laser polishing	714-939-3247
Lawrence D. Weaver International Laser Systems P.O. Box 9316 Albuquerque, NM 87119	Optical properties, optical constants, structure, damage diagnostics & instrumentation	505-844-1064
John M. White Dept. of Chemistry University of Texas Austin, TX 78712	Surface science	512-471-3704

<u>NAME</u>	<u>AREA OF INTEREST</u>	<u>TELEPHONE NO.</u>
Charles Wickersham Battelle Columbus Labs 505 King Avenue Columbus, OH 43201	Explosive crystallization sputtering & evaporation processing	614-424-7710
Richard T. Williams Code 6511 Naval Research Lab Washington, DC 20375	Synchrotron radiation with in-situ deposition, analy- sis, laser testing. (NRL/ NBS facility at NSLS).	202-767-4653
Harry V. Winsor AFOSR/NE Bolling AFB, DC 20332		202-767-4931 297-4931 Autovon
Frank Wodarczyk Rockwell International Science Center P.O. Box 1085 Thousand Oaks, CA 91360	Laser desorption surface analysis	805-498-4545

Appendix B



OPTICAL THIN FILM WORKSHOP

21 - 23 APRIL 1982

Wednesday, 21 April 1982 - Taos Room

8:15 - 9:00	Opening Remarks Purpose of Workshop
9:00 - 10:00	Laser Damage/Dr. Guenther
10:00 - 10:30	Break
10:30 - 11:00	Laser Damage
11:00 - 11:30	Surface Preparation/Dr. Wodarczyk
11:30 - 12:00	Surface Preparation/Dr. Temple
12:00 - 1:30	Lunch
1:30 - 2:00	Surface Preparation/Dr. Knotek
2:00 - 2:30	Surface Dynamics/Dr. Doll
2:30 - 3:00	Thin Film Microstructure/Dr. Messier
3:00 - 3:30	Break
3:30 - 4:00	Thin Film Microstructure/Dr. Sanders
4:00 - 4:30	Explosive Crystallization/Dr. Wickersham
4:30 - 5:00	Amorphous Optical Coatings/Dr. Pawlewicz
5:00 - 5:30	Acoustic Wave Thin Films/Dr. Lakin
7:00 - 9:00 pm	Closed Government Sessions if Required in Nambe Room

OPTICAL THIN FILM WORKSHOP

21 - 23 APRIL 1982

Thursday, 22 April 1982 - Tesuque/Zuni Room

8:00 - 8:30	Thin Film Deposition/Dr. Donovan
8:30 - 9:00	Thin Film Deposition/Dr. Brinker
9:00 - 9:30	Thin Film Deposition/Dr. Rona
9:30 - 10:00	Thin Film Deposition/Dr. Barbee
10:00 - 10:30	Break
10:30 - 11:00	Surface Diagnostics/Dr. Nagel
11:00 - 11:30	Surface Diagnostics/Dr. Merrill
11:30 - 12:00	Surface Diagnostics/Dr. Lindau
12:00 - 1:30	Lunch
1:30 - 5:30	Brainstorming Session in Regent Suites

OPTICAL THIN FILM WORKSHOP

21 - 23 APRIL 1982

Friday, 23 April 1982 - Tesuque/Zuni Room

8:30 - 9:00 Group 1 Report

9:00 - 9:30 Group 2

9:30 - 10:00 Group 3

10:00 - 10:30 Break

10:30 - 11:00 Group 4

11:00 - 11:30 Group 5

## GROUP MEMBERS AND AREAS OF CONCERN

### Group 1

#### Methods for Control of Microstructure

Joe Apfel  
Troy Barbee  
Dick Hoffman  
Myron Maclin  
Bob McNeil  
Russell Messier - Chairman  
Melmet Rona  
Dave Sanders  
Chuck Wickersham  
Harry Winsor

### Group 2

#### Structure of Thin Films

Hal Bennett  
Milt Birnbaum  
J. D. Doll  
Terry Donovan  
Sam Holmes  
Mike Jacobson  
Ken Lakin  
Keith McDowell  
Lee Myers  
Vaidya Nathan  
Walt Pawlewicz  
Earl Rudisill  
Howard Shanks - Chairman

### Group 3

#### Microanalytical Diagnostics

Walt Haas  
Ingolf Lindau - Chairman  
Tilak Raj  
Bob Springer  
Charter Stinespring  
Mike White

Group 4

Substrate Surface Preparation and Cleaning

Richard Esposito  
Bob Merrill - Chairman  
Yoshihara Namba  
R. C. Pastor  
Bruce Pierce  
Paul Temple  
Frank Wodarczyk

Group 5

Structure/Optical Properties that Relate to Damage

Art Guenther  
Ron Lusk  
Jack McIver - Chairman  
Dave Milam  
Larry Weaver

## Appendix C

LASER INDUCED DAMAGE IN OPTICAL THIN FILMS

Dr. Arthur H. Guenther

Air Force Weapons Laboratory

Kirtland Air Force Base

New Mexico 87117

UNITED STATES

## LASER INDUCED DAMAGE IN OPTICAL THIN FILMS

Dr. Arthur H. Guenther

Air Force Weapons Laboratory

Kirtland Air Force Base

New Mexico 87117

UNITED STATES

The motivation for laser damage research should be quite obvious to all individuals working with high power lasers, [1] since: LASER INDUCED DAMAGE IS THE PRINCIPAL LIMITING CONSTRAINT IN THE DESIGN AND OPERATION OF HIGH POWER LASER SYSTEMS RESULTING FROM PERFORMANCE DEGRADATION DUE TO THE INTERACTION OF INTENSE COHERENT LIGHT WITH OPTICAL ELEMENTS IN THE SYSTEM. It is not only a concern in the operation of high power laser systems but is equally important in the design of efficient lasers. If one could handle greater power densities (watts per square centimeter) of laser radiation one could then build more compact, therefore more efficient, lighter weight systems which, for many applications, are a distinct advantage.

Laser damage does not have to be catastrophic in nature. As shown in Figure 1, for CW lasers, a noncatastrophic damage would be the distortion of an optical element which might lead to beam spread while, conversely, a catastrophic CW failure would be the actual fracture of a window due to the energy deposited by the passage of a laser beam. In the pulsed laser case, one might have small scale self-focusing resulting in beam breakup as an example of noncatastrophic damage. While avalanche ionization leading to electrical breakdown would be the most characteristic of the numerous catastrophic types of failure, in the final analysis laser damage occurs when an element no longer adequately performs the function it was designed to perform in the system.



## LASER DAMAGE

MODE OF LASER OPERATION	NON CATASTROPHIC	CATASTROPHIC
CW	THERMAL DISTORTION	THERMAL FAILURE (FRACTURE)
SHORT PULSE	SMALL SCALE SELF FOCUSING	AVALANCHE IONIZATION

Figure 1. Example of catastrophic and noncatastrophic damage for both high energy CW lasers and high peak power pulsed lasers.

In the hierarchy of damage sensitivity optical materials in bulk form would exhibit a damage generally more intrinsic in nature and without question the highest in threshold followed by surfaces and finally the material in thin film form. This change in damage sensitivity can be thought of as proceeding from a fairly intrinsic failure to a more extrinsic failure mode reflecting the influence of imperfections. In many cases the optical electrical field associated with certain power densities corresponds quite closely to the dc fields associated with electrical breakdown of the dielectric. However, in the case of thin films the fields can be as much as two orders of magnitude lower at failure. Surfaces generally fall between these two extremes. They do not have a threshold as high as bulk damage because they are exposed to the environment and they have been usually mechanically processed resulting in stress concentrations, microcracks or other sharp material discontinuities. There is one physical characteristic which changes

drastically as a material proceeds from bulk through surface to thin film form and that is the absorption coefficient. Many materials exhibit absorption coefficients as low as  $10^{-5} \text{ cm}^{-1}$  in the bulk whereas the same material in thin film form may have an absorption coefficient as high as  $10^3 \text{ cm}^{-1}$  to  $10^4 \text{ cm}^{-1}$  which is a nine order of magnitude variation in absorption [2]. This absorption increase is generally attributed to impurities. It is easy to note that surfaces which are midway between the bulk and thin films will also have greater opportunity for contamination and in fact the absorption coefficients of optical surfaces are always higher than bulk values but lower than film values.

For the above reasons it is easy to see that thin films generally form an Achilles heel in high power laser systems and therefore they are the prime candidate for improvement and study. But, before one can adequately study the failure of thin films, one must understand, to some degree, the surfaces of the substrate upon which the films are deposited.

#### EFFECT OF SURFACE ROUGHNESS

One obvious surface quality is roughness. Figure 2 shows how the pulsed laser induced damage threshold of fused silica surfaces vary as the rms surface roughness,  $\sigma = 335$  angstroms. It is gratifying to note (although it may be quite fortuitous) that as you extrapolate the data to a  $\sigma$  value which corresponds to the interatomic spacing in fused silica, the breakdown field approximates the dc breakdown value for quartz. This surface roughness, damage level correlation can be described by the following expression

$$E = \sigma^m = \text{constant} \quad (1)$$

where the value of  $m$  centers around 0.5,  $E$  is the threshold damaging optical field in MV/cm and  $\sigma$  the rms surface roughness in angstroms.

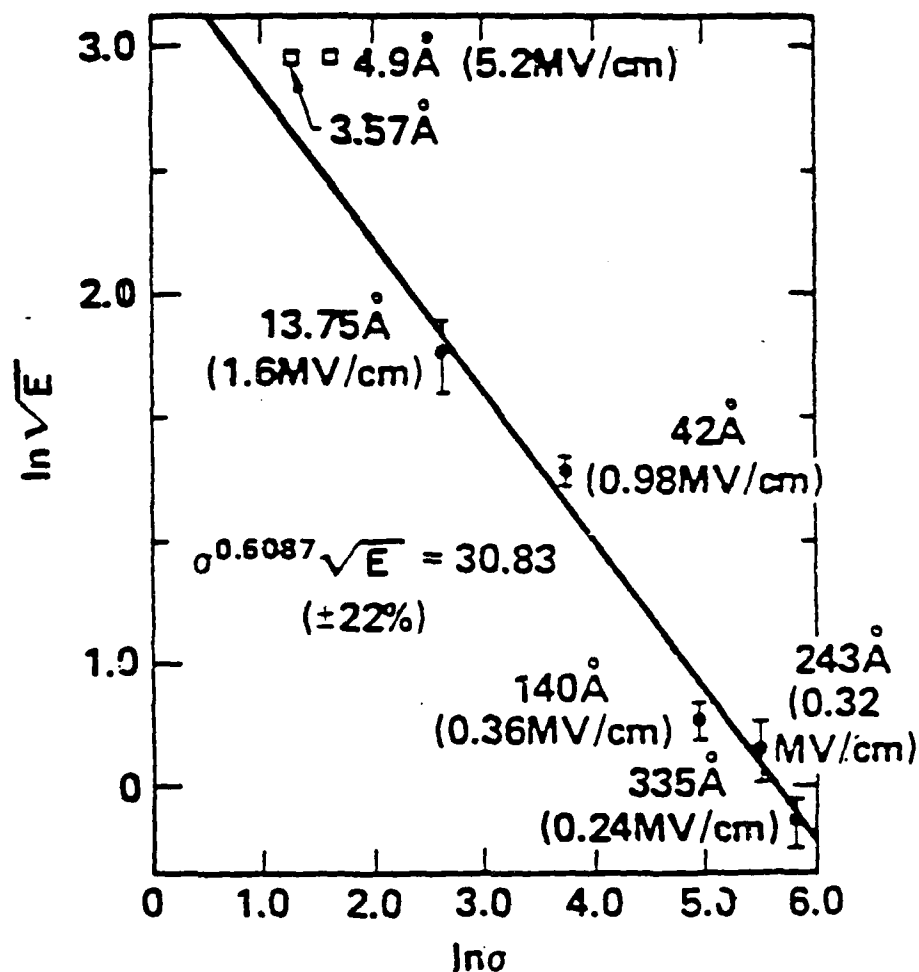


Figure 2. Laser induced damage threshold vs. surface roughness for conventionally polished fused silicon. The laser characteristics were 40 ns FWHM pulse duration 1.06  $\mu$ m wavelength at TEM<sub>00</sub>.

A tentative explanation of this dependence has been given by Babu. [4] Simply stated, he proposed that an increase in exposed surface area per cm<sup>2</sup> of irradiated area will occur as the roughness increases. Thus, there is an increased area upon which impurities can settle leading to an increased probability of damage. He went through a primarily

geometric analysis and arrived at a similar relationship to Equation 1; furthermore the exponent  $m$  was about 0.5 in agreement with the experimental observation. If this simple explanation is correct then surface finishing procedures that would improve the cleanliness of the surface should also lead to an improved damage threshold. As shown in Figure 3, this is the case.

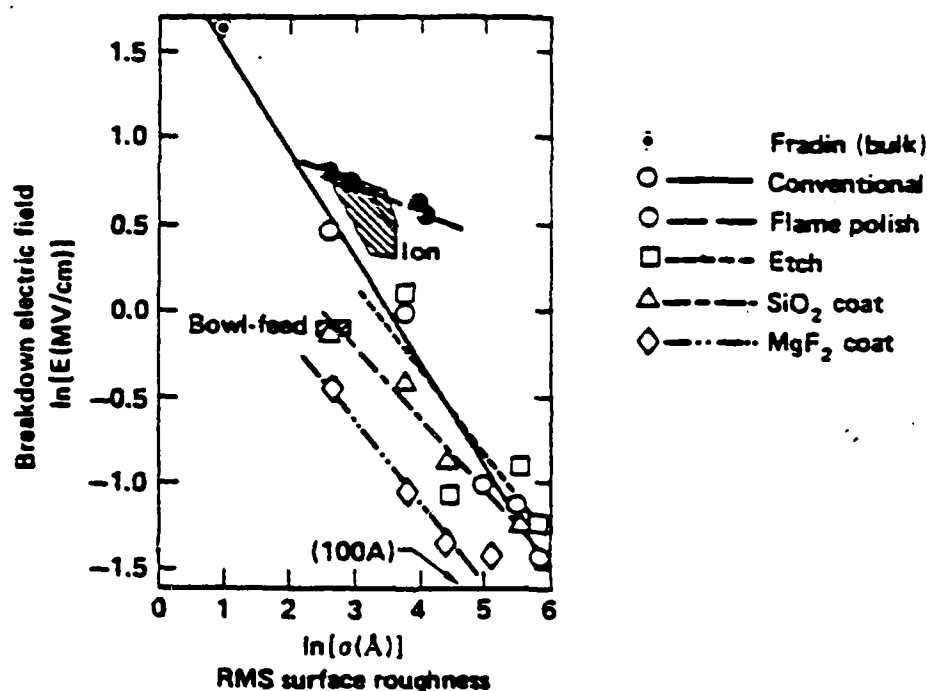


Figure 3. Laser induced damage threshold vs. surface roughness for various surface conditions prepared by different polishing and coating techniques. Experimental parameters were similar to those described in Figure 2.

You will note that acid etched surface damage thresholds are slightly above those for conventionally polished surfaces while ion polishing with helium, xenon, or krypton lead to improved damage thresholds as well. Flame polishing, which is really an annealing of the surface, exhibits improved damage resistance as well at similar  $\sigma$  values.

However, note that the deposition of two thin film coatings, silicon dioxide and magnesium fluoride, led to a decrease in the damage threshold. This would seem to indicate that by coating the material we are not so much covering up the surface irregularities as we are adding impurities or damage initiating sites to the surface.

#### EFFECT OF REFRACTIVE INDEX

After studying the influence of other surface characteristics such as subsurface disorder as well, we ascertained if the above behavior held at different pulse lengths. It was shown that while the general trend, smoother is better, held at shorter pulses the advantage was not evident at picosecond pulse lengths for normally good quality polished optical surfaces. [5] We then embarked upon a study of the film materials themselves.

A word about scaling relationships is in order. Empirical scaling relationships can be very useful in that they allow one to estimate performance under conditions other than where data points are taken. However, one must caution that scaling relationships should not be applied outside the range under which they are validated because the operating mechanisms, that is the physics of interaction, may be different.

It is quite easy to arrive at an adequate range of refractive indices which would cover most normally used coating materials. A scaling relationship was derived which indicated that the damage threshold would vary as:

$$E \approx k/(n^2 - 1)$$

where  $n$  is the refractive index of the film material and  $k$  is a constant dependent upon other material properties. [6] The data for a large number of films is shown in Figure 4. All of the films were

half-wavelength thick at 1.06  $\mu\text{m}$ . For half-wavelength film the optical field is the same at the film/air interface as at the film/substrate interface. Any field differences due to changes in the refractive index are therefore within the film itself.

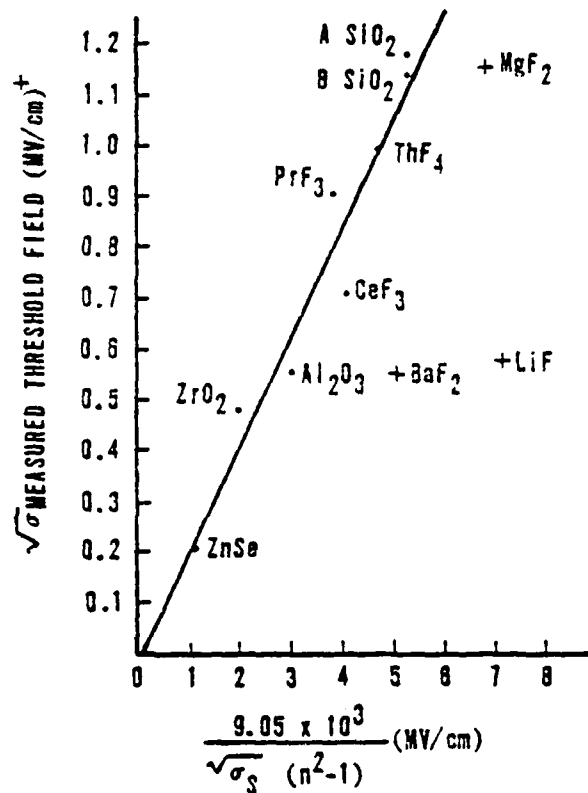


Figure 4. Damage threshold of several half-wave (at 1.06  $\mu\text{m}$ ) thin film coatings on SiO<sub>2</sub>. Films demarked + are inhomogeneous. Roughness corrections have been made assuming a  $\sigma^{0.5}$  ( $\sigma$  = rms surface roughness) correlation. ( $\sigma_s$  is equal to the interatomic spacing;  $n$  is the refractive index. A, silicon dioxide on quartz, B, silicon dioxide on BK-7 substrate.

One should note that we have taken into account the surface roughness of the substrate in calculating the threshold field damage. It is noted that as one proceeds down in index from materials like zinc selenide there are three films which do not seem to behave normally.

They are the fluorides of barium, lithium, and magnesium. We were concerned as to whether our scaling analysis was correct for we knew no reason why these films did not exhibit much higher damage thresholds. Realizing the importance of full characterization of the process variables and film properties associated with the samples, it was easy to ascertain that these three films exhibited a high degree of inhomogeneity as attested by spectro-reflection measurements. The other films all exhibited homogeneous behavior.

To prove that our scaling relationships were correct we repeated the damage assessment experiments on bare surfaces of a large variety of optical materials including crystalline samples of the fluorides of lithium and magnesium. You will note from Figure 5 that the scaling

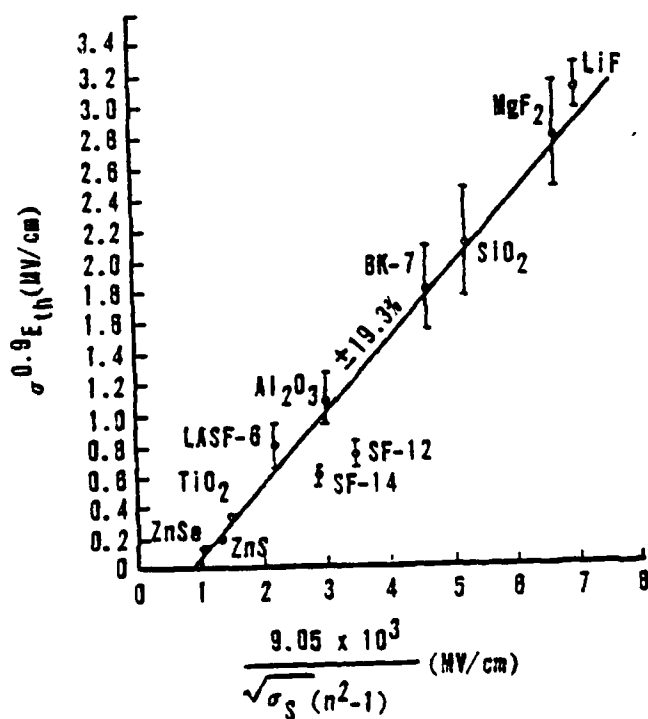


Figure 5. Surface damage threshold of several optical glasses as a function of their refractive index and interatomic spacing, as in Figure 4. Roughness corrections have been included before.

relationship now holds for all materials including lithium and magnesium fluoride over the range of refractive indices studied. This curve depicts the well-known observation that lower index materials are more difficult to damage than higher index materials. Through the use of an appropriate film design and material choice, the production of more damage resistant films can result.

#### OTHER SCALINGS

Scaling with pulse length, spot size, etc. has been accomplished to some degree and is summarized in several articles [7] For a complete description of the scaling of laser damage one is referred to the series of Proceedings of the Symposium on Optical Materials for High Power Lasers, the Boulder Damage Symposium. [8, 9]

#### FREQUENCY SCALING

The most recent scaling study accomplished on thin films has been concerned with their frequency dependence on damage, a most important subject considering the rapid increase in the number of short wavelength lasers whose progress is being impeded by the lack of damage resistant optical elements.

If, in fact, the prime culprit in damage is defects, i.e., impurities, defects, dislocations, voids or whatever imperfection, we might take a lead from some early work in the impurity damage area due to Hopper and Uhlmann. [10] Over a decade ago the major problem facing the glass laser physicist was platinum inclusions. The analysis performed by Hopper and Uhlmann and later by Bennett [11] suggested that platinum



inclusions in the glass host were heated resulting in a thermal fracture of the surrounding medium. In their analysis the impurity particles were considered to be metallic. Their analysis assumed a constant and uniform temperature throughout the impurity which is not too unrealistic considering the high scaling of damage with laser pulse length which is  $\sqrt{t}$ , where  $t$  is the laser pulse width. However, in our case for dielectric thin films, the impurities themselves are primarily dielectric in nature. This can lead to some changes in analysis and predictions. Figure 6 shows the curve of the damage level (in  $\text{J}/\text{cm}^2$ ) versus particle size for a metallic impurity in a dielectric host. The second curve is due to a complete thermal treatment which includes the

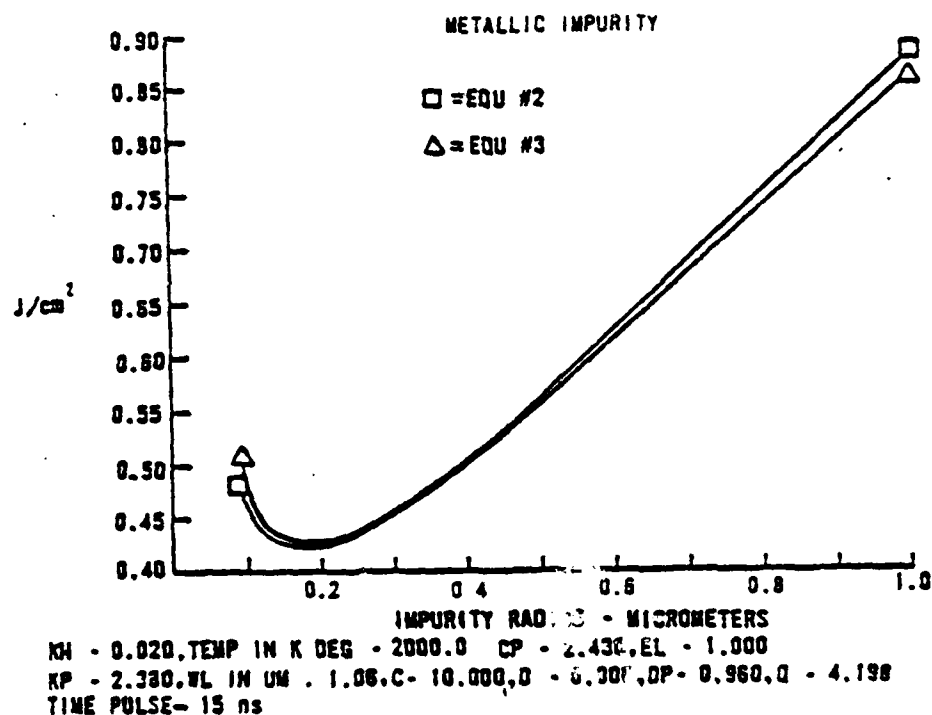


Figure 6. Comparison of the exact solution of thermal equation with approximate solution.

impurity thermal conductivity as a parameter. The full treatment is derived from some early work of Goldenberg and Tranter in the early 1950s. [12] This early work was evidently unknown to the laser damage community and we have been laboring under approximate solutions when, in fact, exact solutions were available. Figure 7 shows the divergence of the two approaches when describing the behavior for a dielectric impurity in a dielectric host where the thermal conductivities of the two are comparable.

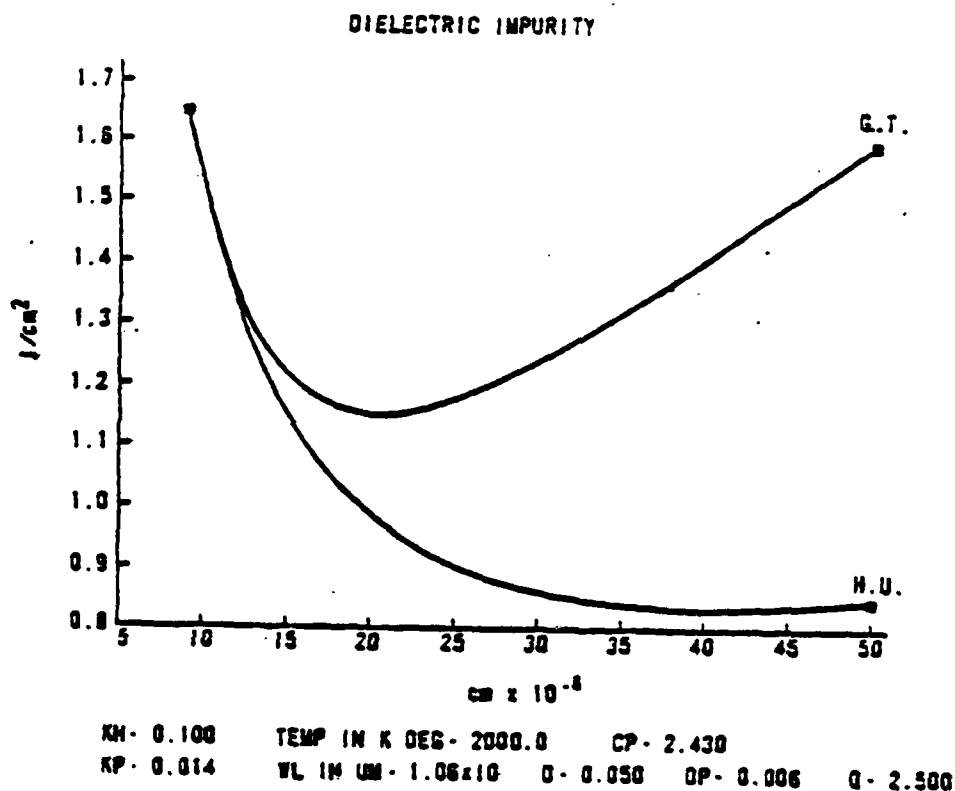


Figure 7. Comparison of the exact solution of thermal equation with approximate solution.

The criteria for damage that we have applied is as follows. When the temperature of the impurity particle arrives at a temperature comparable to the melting point of the host, damage will ensue instantaneously. This criteria is fairly insensitive to the exact temperature since the absorption of laser radiation increases rapidly in these materials at very high temperatures.

To determine if, in fact, this was the appropriate description, we studied ten thin film materials, primarily oxides and fluorides, whose absorption edges varied from 113 nm in the case of magnesium fluoride to about 300 nm in the case of titanium dioxide [13].

We chose this range of absorption edges to allow the additional study of possible multiphoton effects at short wavelengths. Our tests were performed at one micron for a variety of film thicknesses chosen such that we would have half wave films at each of the different wavelengths employed in the testing. This latter feature was indeed fortuitous in that it gave us added proof that our analytical predictions adequately described the experimental results.

A typical set of data is shown in Figures 8-11. The theoretical lines on these curves are due to the application of Goldenberg and Tranter predictions. You will note that if one moves to thinner films the damage sites at shorter wavelengths. Some morphology attributable to multiphoton effects were also seen at the shortest wavelengths. Some morphology attributable to multiphoton effects were also seen at the shortest wavelengths for the longer absorption edged materials, that is the oxides.

Additional support for the impurity dominated failure model can be obtained from a study of the pulse length dependence on failure. If one performs numerous calculations to find the minimum size impurity

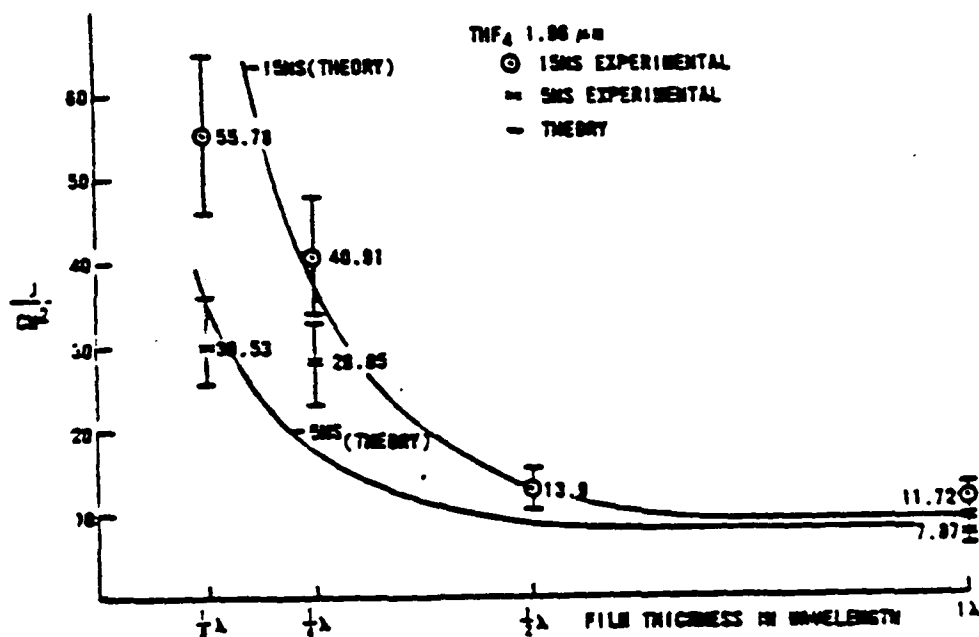


Figure 8. Damage threshold versus film thickness at  $1.06 \mu m$  laser wavelength.

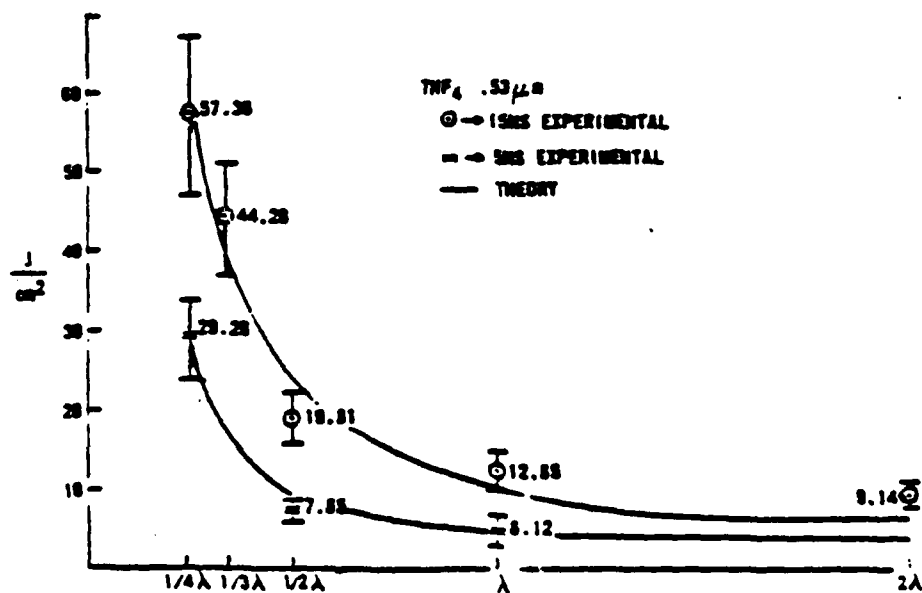
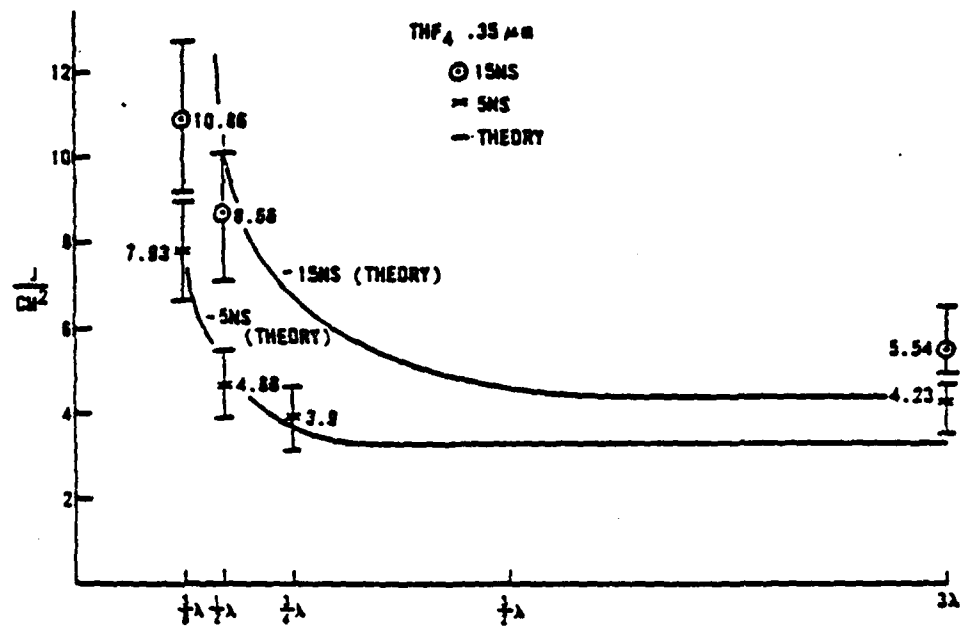
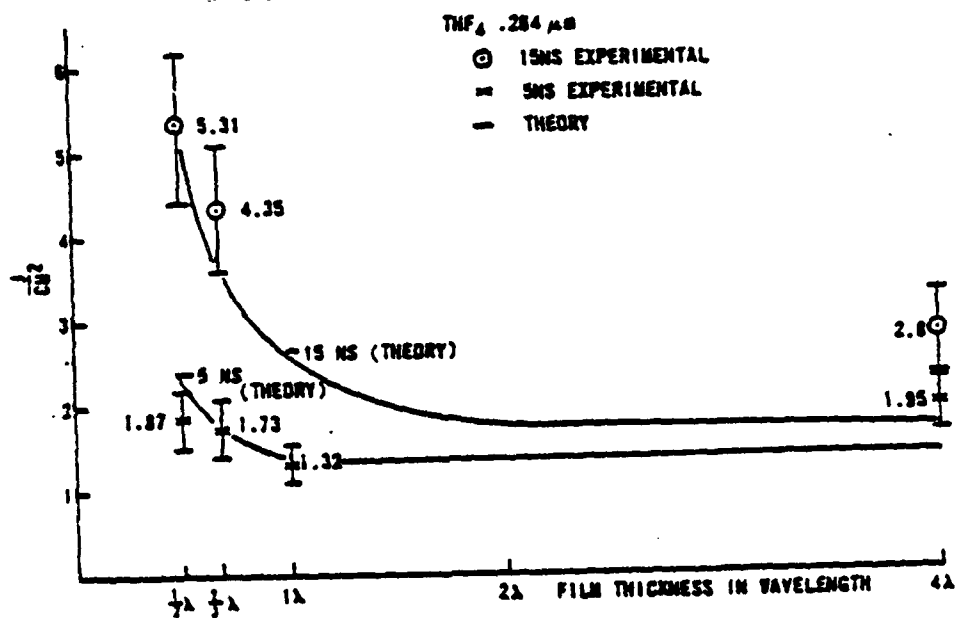


Figure 9. Damage threshold versus film thickness at  $0.53 \mu m$  laser wavelength.



-Figure 10. Damage threshold versus film thickness at 0.35 μm laser wavelength.



-Figure 11. Damage threshold versus film thickness at 0.26 μm laser wavelength.

which will damage at a given pulse length, one can plot a curve of damage versus length. Applying this to some previous work of Milam at Lawrence Livermore National Laboratory [14] on bare fused silica surfaces we are able to quite adequately describe this observation as shown in Figure 12 which covers damage measurements from the subnanosecond to the several nanosecond range. The dependence is not that dissimilar to the  $\sqrt{t}$  dependence although it is a slight improvement in predicting the observed pulse length dependence.

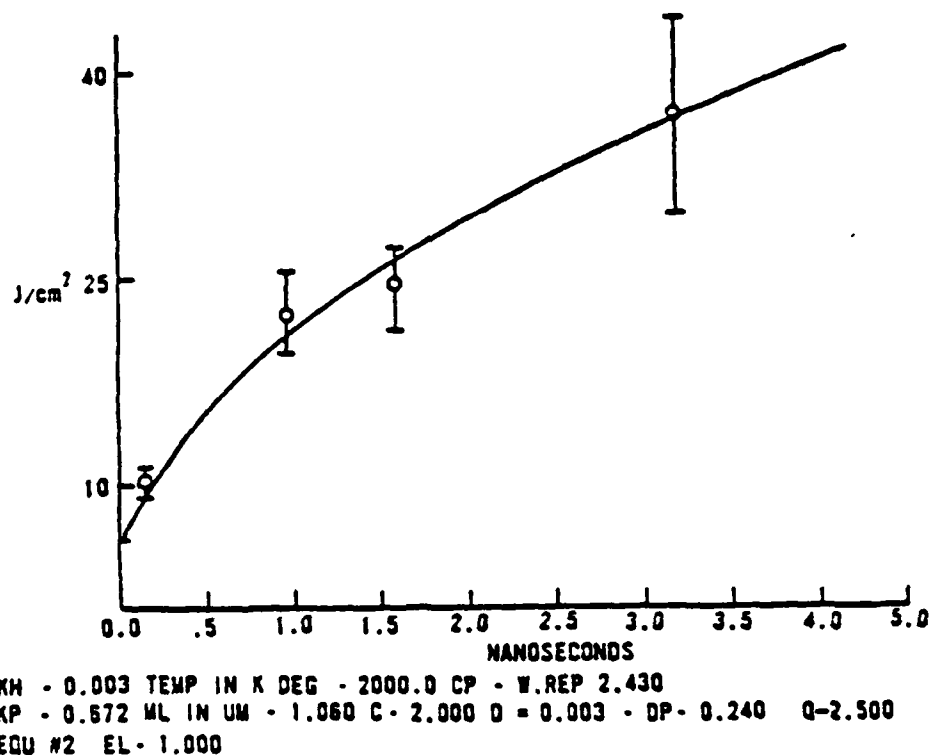


Figure 12. Damage threshold of fused silica surfaces as a function of laser pulse length. The solid curve is theory.

## SUMMARY

Perhaps the reason thin films are the most damage sensitive form of optical materials is because they are the most complex. There are many parameters that are involved in the processing and fabrication of thin films. To improve thin films to the same degree we have improved solid transparent optical materials and to a lesser degree the fabrication of optical surfaces, one must adequately characterize the process variables and the material properties such as its purity, etc. By process variables I mean deposition rate, background gas, pressure, the substrate, temperature, angle of incidence, etc. This is the first step in an optimum approach for progress in optical thin films as shown in Figure 13.

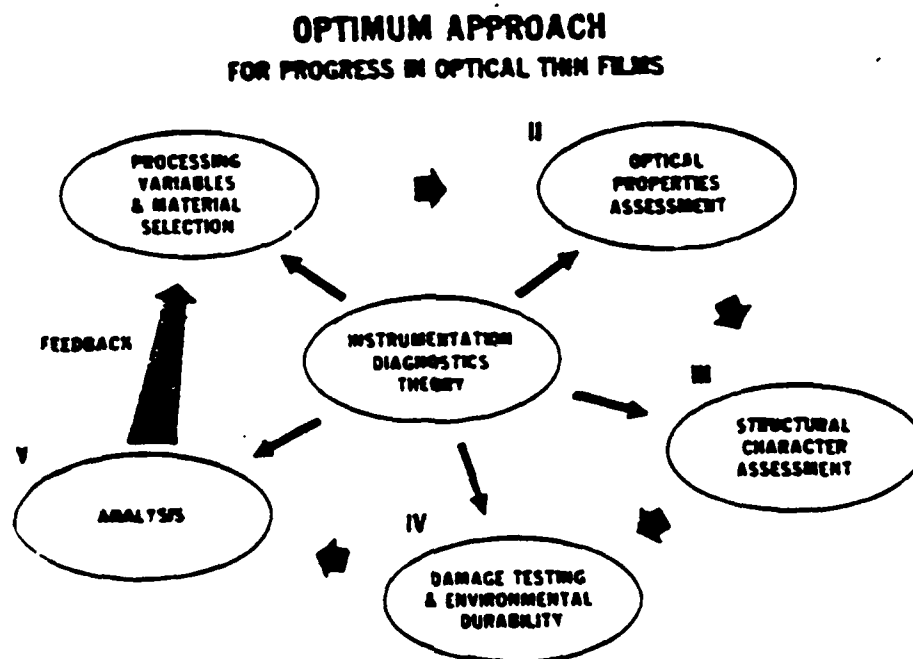


Figure 13. Optimum approach for progress in improving optical thin films.

Following this initial step one would perform careful assessment of the film, optically speaking, how well it performs. This performance must then be correlated with the film structure and that structure related back to the process variables. That is, what is the structure of the film, how it relates to the optical properties achieved by the particular process employed. One then needs to do damage testing to study the environmental degradation to determine which of the many variables are important and then after analysis give feedback to the process. The key to this whole area is characterization and diagnostic instrumentation which is a very expensive situation. However, because of the importance of this subject, I think we may be seeing more of this type of analysis in the future.



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## LASER DESORPTION OF SURFACE CONTAMINANTS

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Experiments involving excimer laser irradiation of various substrates showed that thin metal films could be deposited in the laser-illuminated area. These films formed on both absorbing and nonabsorbing substrates at room temperature with a measurable increase in adhesion over thermally evaporated films. Subsequent experiments using residual gas analysis under ultra-high vacuum conditions show that desorption of surface contaminants, which normally inhibit film nucleation and growth, resulted from absorption of the laser radiation by the contaminating layers. A contaminating layer was found to build up with time, even in a UHV chamber at pressures less than  $10^{-9}$  torr. Laser powers sufficient for surface cleaning of the nonabsorbing substrates were on the order of  $100 \text{ mJ/cm}^2$ .

\*Research funded in part by AFOSR contract F49620-81-C-0074.

CO<sub>2</sub> LASER POLISHING OF FUSED SILICA SURFACES FOR INCREASED  
LASER DAMAGE RESISTANCE AT 1064 nm

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When mechanically polished fused silica surfaces are heated with continuous wave CO<sub>2</sub> laser radiation, a sharp transition from laser-damage-prone to highly laser-damage-resistant surfaces takes place over a relatively small range in CO<sub>2</sub> laser power. 1.06  $\mu$ m, 9 nsec small-spot damage measurements show that some CO<sub>2</sub> laser-treated surfaces have a local damage threshold as high as the bulk damage threshold of SiO<sub>2</sub>. On some CO<sub>2</sub> laser-treated surfaces, 1.06  $\mu$ m, 1 nsec large-spot damage thresholds were increased by a factor of 3 to 4 over thresholds of the original mechanically polished surface. These treated parts show no obvious change in surface appearance, as seen in bright-field, Nomarski, or total internal reflection microscopy. They also show little change in transmissive figure. Of particular interest to this meeting, anti-reflection films deposited on CO<sub>2</sub> laser-treated surfaces have thresholds greater than the thresholds of antireflection films on mechanically polished surfaces.

In this talk I will discuss what has been learned about CO<sub>2</sub> polished surfaces to this time and what we hope to accomplish this fiscal year in a program sponsored by LLNL.

STUDY OF HYDROGEN AND HYDROXYLS ON SURFACES  
BY STIMULATED DESORPTION†

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ABSTRACT

Over the past few years, Electron- and Photon-Stimulated Desorption (ESD and PSD) have been shown to be particularly sensitive to hydrogen on surfaces. Using ESD/PSD it is possible to obtain detailed electronic and structural information on hydrogens bonding site and the nature of hydrogen chemistry on surfaces. Work on Ti and Si and their oxides have shown that several hydrogen species are present, including hydroxides, hydrides and hydrogen bonding to lattice oxygen. Some results and implications of these studies will be presented including a recently discovered radiation sensitivity of  $\text{SiO}_2$  surfaces due to the presence of OH species.

†This work performed at Sandia National Laboratories supported by the U.S. Department of Energy under contract number DE-AC04-76DP-00789.

THEORETICAL STUDIES OF SURFACE DYNAMICS:  
SURFACE SELF-DIFFUSION

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ABSTRACT

Surface diffusion is an important elementary event in many surface processes. Detailed measurements of surface diffusion constants on well-defined substrates have been made through field ion microscope techniques. These diffusion constants exhibit marked dependence on the surface morphology. We examine surface diffusion using recently developed molecular dynamics techniques in an effort to see the extent to which such techniques can quantitatively describe the diffusion process. We also point out a common misconception concerning the nature of adsorbate-adsorbate interaction potentials and their extraction from field ion microscope data.

## MICROSTRUCTURE-PROPERTY RELATIONS IN THIN FILMS

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Probably all physical properties of thin films depend at least in part on their microstructure. A structure zone model common to all vapor deposition methods has been developed to classify the various physical microstructures found [1, 2]. In this model the microstructure is related to a normalized temperature,  $T/T_m$  where  $T$  = temperature of the film during preparation and  $T_m$  = the melting point of the material ( $^{\circ}\text{K}$ ). Thornton [2] extended this model to the case of sputtered films in which a second important variable, the pressure of the sputtering gas (e.g.,  $P_{Ar}$ ) was demonstrated. It has been shown that the densest, most crystalline structures are obtained at high  $T/T_m$  and low  $P_{Ar}$ .

Recently we have shown the effectiveness of this model in understanding the complex relations between chemical, mechanical, optical, and electronic properties and the detailed physical structure for the amorphous semiconductors a-Ge [3], a-Si [4], and a-Si:H [5] prepared by rf-sputtering. In particular, the effects of positive ion-bombardment of the growing film have been shown to be most important [5] and directly related to decreasing gas pressure. In effect, the bombardment is producing a second source of temperature, albeit a non-equilibrium temperature, at the growing film. Thus the "effective  $T/T_m$ " can be increased without increasing the temperature of the workpiece. This process is seen to be especially important for material with a high  $T_m$  (e.g., ceramics) since it allows dense structures to be achieved at relatively low equilibrium, external heater temperatures.

In addition to reviewing this above work [3-5] in the context of general deposition processes, we will present recent unpublished results on SiC and BN films in which similar bombardment effects are related to epitaxial growth and film hardness. Relations between our work and that reported for optical coatings prepared by any technique will be made. A general, evolutionary model of film growth will be presented. Finally, methods we have used for characterizing the physical structure of thin films in detail, from the micron-level (micro-structure) to the atomic-level (nanostructure), will be described.

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## AMORPHOUS THIN FILMS FOR OPTICAL APPLICATIONS

D. M. Sanders

F. N. Farabaugh

W. K. Haller

Glass has long been the preferred state for bulk optical components because of the high transparency possible. Recently, there have been indications that glassy thin films may also be superior for optical applications when compared to their polycrystalline counterparts. This talk outlines the general considerations which lead to bulk glass formation. After this introduction, we discuss thin film glass formation using Multisowne electron beam co-deposition in the systems  $\text{ZrO}_2 - \text{SiO}_2$  and  $\text{Zr}_2 - \text{MgO}$ . It was found that x-ray amorphous coatings could be produced on heated substrates using co-evaporation. From this we conclude that it is not necessary to use traditional glass formers to produce glassy coatings. The use of mixtures is sufficient.

## EXPLOSIVE CRYSTALLIZATION OF AMORPHOUS MATERIALS

Charles E. Wickersham

Explosive crystallization is a rapid amorphous to crystalline phase transformation which occurs in certain amorphous materials when a localized energy impulse (optical, electrical or mechanical) interacts with amorphous layers. The phenomenon is easily recognized by the rapid propagation of a crystallization front, which once initiated at the localized impulse point propagates radially through the amorphous layer. A number of changes in the layer properties occur as a result of the amorphous to crystalline transformation. These property changes can be exploited in a number of applications. In addition, it is possible that explosive crystallization type phenomena may be a contributing factor in the laser damage of *amorphous materials*. Previous work in this field will be reviewed and recent experimental results with amorphous germanium layers will be described which support an energy balance model for this phenomenon.

## AMORPHOUS OPTICAL COATINGS: THE OPTIMUM COATING MORPHOLOGY?

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### ABSTRACT

Research at Pacific Northwest Laboratory during the past few years with reactively sputtered optical coatings has resulted in a significant body of information which suggests that amorphous or glassy optical coatings are superior in properties to polycrystalline coatings. Amorphous  $\text{TiO}_2$  coatings were found to have damage thresholds twice as high as polycrystalline  $\text{TiO}_2$  coatings in Nd:glass fusion laser tests with 1 nsec pulses at 1.06 microns wavelength. The amorphous  $\text{TiO}_2$  coatings have extremely smooth surfaces which result in very low scatter compared to typical polycrystalline "cobblestone" topography. Surface roughness and scattering are found to increase in direct proportion to grain size. Similar results have been obtained for amorphous  $\text{Ta}_2\text{O}_5$  and  $\text{Nb}_2\text{O}_5$ . Record low absorption has been measured for amorphous Si:H alloy coatings in the near infrared (1 to 3 microns). The Si:H coatings, like the oxide coatings, are very low in scatter. Their surface roughness does not increase appreciably with coating thickness. Implications of these results and directions for future research are discussed.

Work supported by the Materials Sciences Division of the Office of Basic Energy Sciences, U. S. Department of Energy.

†Operated by Battelle Memorial Institute for the U. S. Department of Energy under contract DE-AC06-76RLO 1830.

## ACOUSTIC WAVE THIN FILMS

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This paper presents a perspective on thin films that is different from those associated with optical coatings. In order to present this perspective, the following topics are discussed: applications, material requirements, growth methods, and evaluation techniques.

The applications of thin films in acoustics primarily involves transduction in mechanical resonators or delay line signal processing devices. For bulk wave transduction, the film's crystal symmetry must be controlled in order to selectively excite the desired propagation mode. Most difficult are c-axis in-plane films for shear wave generation.

Material requirements are possibly more severe than those encountered in optical coatings. Films must be piezoelectric, have high  $Q$  (greater than 1,000 at 1 GHz), exhibit long term stability of properties measured in parts per million, and be compatible with various processing techniques. In general, attempts are made to obtain these properties in epitaxial or highly oriented films.

Growth methods are: chemical vapor deposition, vapor transport, sputtering, and molecular beam epitaxy. Results for ZnO and AlN films will be presented.

Evaluation of thin films involve optical and scanning electron microscopy,  $\theta$  and  $2\theta$  x-ray diffraction analysis, reflection electron diffractometry, Auger analysis, chemical stability tests, and macroscopic measurements of electromechanical parameters. The microscopy techniques are used to evaluate the film morphology and microcracking.

X-ray diffraction is used to give a quantitative measure of crystal film perfection. Auger analysis of AlN has shown the oxygen content of CVD grown films to be less than 0.5%. Electrical measurements on AlN thin film resonators has shown Q's greater than 1,000 and electromechanical coupling coefficients greater than 10% for resonant frequencies above 1 GHz.

Basic research needs in the area of thin film growth in CVD and sputtering systems is also discussed. Clearly the fundamental growth mechanisms are common to all thin film technologies and a better fundamental understanding would be mutually beneficial.

## ION BOMBARDMENT AND SPUTTER DEPOSITION OF OPTICAL COATINGS

T. M. Donovan, Naval Weapons Center

Absorption losses in evaporated thin films and multilayer structures designed for use in the 3 micron region usually relate to water contamination at interfaces, surfaces, and at localized defects.

An ion beam technique, resonant nuclear reaction analysis, and IR calorimetry have been used to determine concentration and spatial distribution of water in the films.<sup>1</sup> Mass analysis of gas desorbed from localized surface sites by a pulsed HF/DF laser beam is useful for detecting occluded contaminants which reduce laser damage thresholds.<sup>2</sup>

These diagnostics along with ion bombardment and sputter deposition processes designed to minimize defects and contamination in the films will be discussed in this paper. The properties of multilayer enhanced reflectance mirrors deposited by reactive sputtering will also be described.

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## SOL-GEL DERIVED COATINGS<sup>†</sup>

by

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### ABSTRACT

We have used sol-gel process to apply coatings to glass, ceramic, and metallic substrates. The particular process we employed uses metal alkoxides of network forming cations (e.g., Si, Ti, Al, B) as glass precursors. In alcoholic solutions catalyzed by additions of acid or base, the alkoxides are partially hydrolyzed and polymerized to form glass-like networks cross linked by bridging oxygens. Prior to gelation, solutions are diluted and applied to substrates by conventional dipping, spinning, or spraying. The coatings are densified by moderate heat treatments at or below the glass transition temperature. We have developed antireflective, protective, and dielectric coatings employing the sol-gel process. These coatings are comparable optically to chemically vapor deposited coatings and have the advantages of easily controlled stoichiometry, low processing temperatures, and minimum equipment cost.

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<sup>†</sup>This work performed at Sandia National Laboratories, supported by the U.S. Department of Energy under contract DE-AC04-76D099789.

\*A U.S. DOE facility.

## MOLECULAR BEAM EPITAXY

Mehmet Rona

MBE is a technique for the deposition of single crystal films in ultra high vacuum. The technique has been developed within the last five to ten years predominantly by the semiconductor industry.

In vacuum of approximately  $10^{-10}$  Torr, materials are thermally evaporated from boron nitride crucibles onto a substrate, the temperature of which is precisely regulated. Single crystal films up to 5 cm OD have been grown that demonstrate surface roughness no worse than one atomic layer locally (approximately 3-5Å) and films of different chemical compositions but similar lattice structures have been deposited on one another with precisely defined chemical boundaries and no inter-layer diffusion.

Multilayer dielectric coatings for high energy laser system have consistently failed to provide performances expected on the basis of bulk materials properties. Instead, the observed effects support the following simplified model of the behavior of dielectric materials in the presence of intense electric fields.

Polycrystalline structures, with large surface-to-volume ratios, lead to the creation of a high density of surface energy states within the forbidden energy gap of the dielectric and thus to the easy creation of excess electrons in the conduction band under the influence of incident photons.

Electrons within the conduction band are accelerated and transfer their energy easily and rapidly to the lattice because of the polycrystalline nature of the film so that melting and breakdown occur at lower intensities than would otherwise be expected.

The substitution of polycrystalline films by single crystal films should offer improvement on two counts: (a) reduced absorption of energy by electrons in low energy surface states, and (b) longer mean free paths of electrons in the conduction band, with reduced possibility of electron avalanche breakdown and massive energy transfer to the lattice.



## DEPOSITION PHASE DIAGRAMS

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The dependence of film properties on the intrinsic characteristics of the species deposited, the synthesis process used, the resulting structure of the deposited film, the basic characteristics of the synthesis environment, and substrate character are incompletely defined for almost all processes and materials which have been experimentally investigated. In only a few cases has fairly complete work been reported which yields a reasonable view of these mutual interdependencies, with extrapolation to application basically absent.

In this talk, a short review of work in which deposition rate, substrate temperature, and substrate character effects have been specifically determined is given. Such structures have been almost wholly directed to elemental deposits and processes in which extreme care to eliminate cleanliness issues have been taken. In a real sense such results result in a deposition phase diagram.

In this talk this concept of a deposition phase diagram will be expanded upon. Particular emphasis will be on experimental synthesis processes which can be adopted for such explanations and the types of results which can be obtained. Multicomponent systems will be described and survey type experiments discussed. In this case both reactive type phase spread and dual evaporate source experiments are described.

## CHEMICAL AND STRUCTURAL CHARACTERIZATION

### OF $\text{ThF}_4$ LASER-MIRRORS COATINGS

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#### ABSTRACT

A variety of techniques is being employed to determine the chemical composition and atomic structure of thin  $\text{ThF}_4$  films in order to understand variations in the optical reflectivity of laser mirrors coated with this material. Rutherford backscattering of ions and x-ray emission excited by both ion impact and absorption of synchrotron radiation yielded data on impurity concentrations. A relationship between Ta levels and the  $3.8 \mu\text{m}$  absorption coefficient will be reported. X-ray diffraction, in a Read camera using film recording and in a conventional diffractometer with electronic recording, is yielding structural information. Radial distribution functions are obtained from Fourier analyses of the diffractometer scans. The structure of the  $\text{ThF}_4$  films is being studied as a function of annealing temperatures which simulates in-service laser heating. Other techniques available at NRL for analysis of laser-mirror coatings will be described briefly.

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INVESTIGATION OF OPTICAL THIN FILMS ON AN ATOMIC  
SCALE USING SYNCHROTRON RADIATION

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In this talk we illustrate with a few examples how some of the unique capabilities of synchrotron radiation can be used to study optical thin films. The possibility to determine the structural and electronic properties of thin films on an atomic scale will be emphasized, in particular the bonding between the atoms in the thin film and the bonding to the substrate.

Results for the electronic and geometrical structure of Cu overlayers on Pt will be presented. The electronic properties of thin Cu overlayers embedded in Pt depend on the crystal orientation of the Pt substrate. The nature of the Pt-Cu bond, mainly studied by photoemission from the Cu third states, depends on the local environment. The evolution of the properties of the Cu adlayer from isolated atoms to bulk metal can be followed.

The EXAFS technique (Extended X-Ray Adsorption Fine Structure) using synchrotron radiation is now a well-established technique for structural determinations. Recent advancements of the technique have made it possible to determine metal-metal bonding distances in small clusters and follow the changes in these distances from clusters to bulk metal films. Results for Cu and Ni clusters will be discussed.

Extensive knowledge is presently available about the bulk electronic properties of metals. Much less is known about how these properties depend on the surface morphology of the film. Two techniques, photoemission and x-ray absorption have been used to probe how the d-electron occupancy (filled and empty, respectively) in transition metals depends on the local arrangement of the surface atoms in a thin metal film.

The chemical bonding between a metal and a semiconductor is of great interest in a large number of fields, for instance silicides (transition metal-silicon). We will use this system as an example to illustrate how detailed information about the metal-substrate bonding can be obtained on an atomic scale. The effect of impurities, in particular oxygen, on the chemical bonding and the abruptness of the metal-substrate interface will also be discussed.

The presence of hydrogen and its effect on the absorption in thin films has been the subject for much discussion. A newly developed technique, photon stimulated desorption, can detect the presence of hydrogen and also provide details about the hydrogen bonding to the host metal/semiconductor. We will illustrate how this technique can be combined with other surface analysis tools to provide a direct correlation between the structural/electronic properties of a thin surface film and the presence of hydrogen.